MONTE-CARLO SIMULATION OF EPITAXIAL GROWTH: GexSi(1-x)/Si INTERFACES
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Abstract. -- Epitaxial growth of Ge\textsubscript{x}Si\textsubscript{1-x}/Si interfaces is simulated employing a direct Monte-Carlo technique. In addition to various inter- and intra-layer diffusion processes of Si and Ge adatoms, we include a model strain-driven mechanism that gives rise to defects (local out-of-registry regions) in the overgrowth. Our simulation indicates that: (i) the overgrowth is defect-free for \( x \leq 0.2 \); (ii) defect-formation is inevitable for \( x > 0.5 \); and (iii) the transition between the two cases occurs smoothly within \( 0.2 < x < 0.5 \).

Successful growth of Ge\textsubscript{x}Si\textsubscript{1-x} alloy films on silicon substrates has been reported using molecular-beam-epitaxy (MBE) \cite{1, 2}. It has been shown that: at growth temperatures \( T_g \) of 400 - 550°C, the growth mode is two-dimensional (layer-by-layer) for the entire range of alloy composition \( x \) (i.e. lattice-parameter misfit up to 4.2%); at higher temperatures, an abrupt transition from two-dimensional to three-dimensional (islanding) growth occurs at a critical composition \( x_c \). Further examination by use of TEM confirms that thick alloy films grown at \( T_g \approx 550°C \) contain inhomogeneous strain and dislocations when \( x \) is relatively large -- clear evidence of a transition from commensurate to incommensurate growth upon increasing the Ge fraction. In view of the wealth of information regarding growth morphology and crystallinity, as well as the observation of long-range order in the Ge\textsubscript{x}Si\textsubscript{1-x}/Si strained superlattice system \cite{3}, questions naturally emerge as to how the strain imposed by the Si substrate plays a role in driving growth kinetics and stabilizing or deteriorating the microscopic structures of the overgrowth. In this paper, we present our simulation results for epitaxial growth of this novel semiconductor system, specifically below \( T_g \approx 550°C \), employing a direct Monté-Carlo (MC) technique \cite{4}. (The simulation of macroscopic islanding above 550°C would be difficult because of the use of finite-size lattices that is inherent in MC; we refer to Ref.\textsuperscript{5} which presents an equilibrium theory that accounts for morphological transitions at high growth temperatures.)

The large number of kinetic rates in our MC simulations are determined as follows. In keeping with the MBE procedure of Bean et al.\cite{1}, Ge and Si atoms are codeposited onto the Si(001) surface (with the number ratio \( x \) versus \( 1-x \)) determined by a random number generator; the time scale is fixed so as to yield 4 layers per second. (The total deposition rate reported in Ref.\textsuperscript{1} is 5Å/sec.) We ignore evaporation processes, and assume an Arrhenius behavior,
R=Ro exp(-E/kBTg), for both inter- and intra-layer diffusion processes. The activation energy E in each process is determined by counting the number of bonds to be broken in our tight-binding model. For example, a Si atom, that is bonded with Si and Ge at the first nearest neighbor sites on the plane one layer below, has to overcome a barrier expressed as:

\[ E_{Si\rightarrow Ge, Si} = A \{ V_1(Si-Si) + V_1(Si-Ge) + B \{ V_2(Si-Si)(1-x) + V_2(Si-Ge)(x)\} \} \]

where \( V_1(V_2) \) is the first(second)-nearest-neighbor bond strength, and, A and B are adjustable parameters. Note that we distinguish the number and type of neighboring atoms only for the first neighbors, and that the virtual-crystal approximation is used for the second neighbors. The values of \( V_1 \) and \( V_2 \) are extracted using the cohesive energies per bond for bulk Si and bulk Ge (\( E_{coh}(Si)=2.32eV, E_{coh}(Ge)=1.94eV \)) [6], and the Harrison's 1/d^2-scaling rule [6]. The resulting values are: \( V_1(Si-Si)=1.09 \), \( V_2(Si-Si)=0.409 \), \( V_1(Ge-Ge)=0.913 \), \( V_2(Ge-Ge)=0.342 \) (in eV). We use arithmetic means for \( V_1(Ge-Si) \) and \( V_2(Ge-Si) \). It is worth noting the important qualitative physics underlying the values of the above determined parameters. The fact that the cohesive energy (and, of course, the melting temperature) of Ge is lower than that of Si dictates that Ge atoms are kinetically much more active than Si atoms (if they coexist below the melting temperature of Ge, which is 958°C). This may be one of the characteristic features associated with GeSi(1-x) growth, and it is explicitly taken into account in our simulations. The relative magnitude of the pre-exponential factor Ro in the cases of Ge bonded with Ge and Si bonded with Si is estimated from molecular-dynamics results [7] that have been obtained using the Stillinger-Weber potential [8]. The order of magnitude is left as an adjustable parameter C:

\[ Ro_{Si\rightarrow Si} = Ro_{Si\rightarrow Si} = 3.9 \times C \] (hops/sec).

Likewise, Ro for the case of Ge atoms is \( 3.1 \times C \). For a configuration that involves both Si and Ge, we use an appropriate geometric mean. In adjusting the parameters A, B, and C, we found that qualitative results obtained using rapid rates and a large lattice can be reproduced if we use slower rates and a corresponding smaller lattice. An important consideration in determining these parameters is that the growth mode must be layer-by-layer even at growth temperatures as low as 400°C and Ge fraction as low as 0.1, as has been reported [1]. Our optimization procedure yielded the values \( A=0.6, B=3 \), and \( C=10^{11} \), for lattice size 20x20 in the x-y plane with periodic boundary conditions.

In considering the effects of strain, we make the reasonable assumption that the stress imposed by the Si substrate essentially acts on a Ge-environment. A planar cluster that consists of more than two Ge atoms would therefore experience a strong compressive force. In order to take such local strain fields into account, we include a process by which a central Ge atom, that is surrounded by an environment containing more than one Ge atom at the neighboring sites on the same plane, undergoes upward hopping to the plane one layer above. Unless subsequent diffusion processes take place to fill the vacancy, the bond lengths of the surrounding Ge atoms would relax so as to recover, to some extent, the natural bond length. The positions of atoms in the region near such a vacancy should therefore be taken as out of registry. (This can occur even at the monolayer or bilayer level.) We interpret this type of vacancy in our simulations as a defect produced by the effect of strain.

In Fig.1, we display our simulation results for growth profiles up to 10 layers for four different growth conditions. In all cases, the overall growth mode is layer-by-layer. In particular, it can be seen clearly in Figs.1(a) and (c) that the quality of the growth front improves as Tg increases, because of the high diffusion rates at high temperatures. We note, however, that there is a fluctuation in the number of incorporated atoms for x=0.5, as the growth time proceeds. This is due to the competition between the rapid diffusion of Ge atoms and the defect formation; the former helps give rise to smooth layer-by-layer epitaxy, whereas the latter deteriorates the
Fig. 1: Number of atoms per monolayer as a function of growth time (in seconds) for: (a) x=0.2, T_g=550°C; (b) x=0.5, T_g=550°C; (c) x=0.2, T_g=400°C; and (d) x=0.5, T_g=400°C.

Fig. 2: Configuration of Si (open circles) and Ge (solid circles) atoms near the vacancy (dashed circle) obtained in our simulation for x=0.5 and T_g=550°C. This is the view along the -z direction ([001] is the growth direction). The smallest (largest) circles refer to the atoms in the first (fourth) layer.
crystallinity. In Fig. 2, we show one of the configurations around a defect region obtained in our simulation for $x=0.5$ and $T_s=550^\circ C$. The total number of defects in the first two layers is 2 and 8 for the cases of $x=0.5$ and $T_s=550^\circ C$ (Fig. 1(b)), and $x=0.5$ and $T_s=400^\circ C$ (Fig. 1(d)), respectively. This number increases as $x$ increases. No defect is found for $x=0.2$ (Figs. 1(a) and 1(c)). In the region $0.2<x<0.5$ and $400^\circ C<T_s<550^\circ C$, our simulations yield one defect in some cases, and none in others. Due to the use of finite-size lattices, it is difficult to provide a quantitative account of the dependence of defect formation on alloy composition and growth temperature. The present simulations, however, indicate that: (i) $x=0.2$ is the definitely-defect-free region; (ii) $x=0.5$ is the region where defect formation in the overgrowth is inevitable; and (iii) the transition between the two cases occurs smoothly within $0.2<x<0.5$. The above qualitative features would be borne out even if the simulations were carried on till macroscopic thicknesses were reached. This is because the creation of a defect is controlled by the probability for Ge atoms to form a cluster: alloy statistics and topology are essential ingredients in this model.

It is interesting to note that the observed critical thickness $h_c$ and residual strain values are far in excess of theoretical predictions [9]. This may be an indication that, in addition to ordinary misfit dislocations, defects (which correspond to small out-of-registry regions in our model) might be important in determining the growth (especially for $x>0.2$). Firstly, in the defect regions, the strain is somewhat released so that a higher $h_c$ can be attained than the case of no defects. Secondly, when the thickness reaches $h_c$, some of the defects may play a role in nucleation of misfit dislocations, whereas the remaining defects, as well as Ge-clusters without vacancies, would retain the unreleased part of the strain, which could be the residual strain that is observed.

In conclusion, we have made simulation studies of the epitaxial growth of Ge$_x$Si$_{1-x}$ alloys on Si substrates. We have accommodated a simple defect model in a direct MC technique to take the effects of local strain into account. A further investigation is being made, using larger lattices and more sophisticated models, to study in detail the strain-driven mechanisms that give rise to incoherency, phase segregation, long-range order, and other aspects of this interesting and technologically important system.

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REFERENCES